Ferromagnetism in itinerant two-dimensional t_{2g} systems

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Motivated by the recent indications of ferromagnetism in transition metal oxide heterostructures, we propose a possible mechanism to generate ferromagnetism for itinerant t_{2g} systems in two spatial dimensions that does not rely on the coupling between local moments and conduction electrons. We particularly emphasize the orbital nature of different bands and show that, when the Fermi level lies near the bottom of the upper bands, the quasi-one-dimensional nature of the upper bands leads to a large magnetic susceptibility, and thus the ferromagnetic inter-orbital Hund's coupling can easily polarize the upper bands and drive the system into a ferromagnetic state. We discuss the connection between our mechanism with several itinerant t_{2g} systems that may have ferromagnetic instabilities.

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Possible ferromagnetism at polar interfaces between $SrTiO_3$ (STO) and other oxides such as $LaAlO_3$ (LAO) or $GdTiO_3$ (GTO)[1–6] has raised considerable excitement. Such ferromagnetism is remarkable as the electrons are believed to reside nearly completely in the t_{2g} bands of the STO, where there are no localized partially filled shells to form local moments. Ferromagnetism in purely itinerant systems, while envisioned long ago[7], is quite rare in practice; most examples may be at least partially attributed to local moments formed by partially filled d-shells (and often other delocalized electrons).

In this paper, we discuss the possibility of ferromagnetism in t_{2g} systems of this type. Since ferromagnetic polarization inevitably costs kinetic energy, due to the Pauli exclusion principle, the key issue is how the kinetic energy can be minimized. This may occur when electrons are close to Mott localization, which requires *high electron density* in the most strict sense, i.e. approaching a substantial fraction of unity *per atom*. Alternatively, kinetic energy can be suppressed by Anderson localization, i.e. disorder; for now we concentrate on clean systems, however. Otherwise, details of band structure may provide reduced dimensionality which also lowers kinetic energy. Such a mechanism of inducing ferromagnetism is much rarer, but is known to occur. An extreme example is the ferromagnetic state at $\nu = 1$ in the quantum Hall regime, in which a completely flat band is produced by the orbital effect of the magnetic field[8].

We would like to make a distinction between ferromagnetism, and correlated electronic phases more generally, caused by *local* or *long-range* Coulomb interactions. Most correlated electron physics occurs in transition metal compounds and is attributed to the former, local Hubbard-like interaction. Since such local interactions are effective only if there would be a substantial probability for uncorrelated electrons to occupy the same site, they lead to correlation effects only at high electron density. Local interactions of this type are almost completely negligible in typical doped semiconductors. By contrast, long-range Coulomb interactions become increasingly important at low carrier density, and dominate the interaction physics in semiconductors. When this is the case, an effective mass description generally applies, and dimensional analysis implies that the important parameter describing the strength of correlations is the dimensionless quantity, r_s, which gives the ratio of the $1/r$ Coulomb energy to kinetic energy per electron. It increases with decreasing electron density, $r_s \sim n^{-1/d}$, in d dimensions. This "jellium" problem has been extensively analyzed by highly accurate quantum Monte Carlo simulations, and it is known that quite large values of r_s are needed to destabilize the ordinary, paramagnetic metallic state ($r_s > 37 \pm 5$ in two dimensions and $r_s > 75 \pm 5$ in three dimensions[9, 10]). Since the corresponding electron density is extremely small, this itinerant ferromagnetism in low density systems due to this Stoner's mechanism is extremely fragile if it exists at all, and we disregard it in the following.

We are led therefore to consider high electron density ferromagnetism. In the aforementioned LAO/STO and GTO/STO interfaces, there is an intrinsic mechanism for high carrier density: the polar discontinuity[11]. LAO and GTO have a structure of polar (001) layers: $La^{3+}O^{2-}$ has a net charge of $+1$ per unit cell, while $Al^{3+}(O^{2-})_2$ has a net charge of -1 per unit cell (the same counting holds for GTO). STO by contrast is nonpolar. At an ideal (i.e. without atomic reconstruction or compensating defects) interface between two such materials, an electron gas is predicted to arise with a carrier density of *half* an electron per two-dimensional unit cell. This translates, using the unit cell of STO, into a two-dimensional carrier density of $n = 3.5 \times 10^{14} cm^{-2}$, which is extremely large by semiconductor standards. But is it high enough for ferromagnetism and correlation physics?

It is important to realize that a large *two-dimensional* density does not necessarily imply a large electron density per atom. Due to the finite width of the confined wavefunction of electrons in STO, which has been estimated

now in many calculations[12–15], the electron occupation *per site* should be less than 20%, even for the Ti sites nearest the interface. This fact alone makes *strong* ferromagnetism hard to understand. Modern computational studies have put strong restrictions on ferromagnetism in Hubbard models[16]. The most recent studies argue that ferromagnetism is absent in the two-dimensional Hubbard model for fractional site occupation $x \lesssim 0.7$, *even when the on-site Hubbard interaction* $U \to \infty$[17, 18].

The remainder of the bulk of this paper explores the possibility of *weak* (i.e. small moment, low temperature) ferromagnetism in t_{2g} systems at carrier densities which are high by semiconductor standards, but still achievable near STO interfaces and in bulk doped STO. In particular, we will assume that the fractional filling of all bands is sufficiently small that only states near the bottom of a band need to be considered, but that the contribution from on-site interactions is not negligible. We will refer to this as intermediate-high carrier density. We first discuss the origin of possible ferromagnetic instability for a (001) LAO-STO interface and then apply the results to other interfaces and materials. The main result of this paper is that, despite the aforementioned results for the single-band Hubbard model, we find that the directional character of the t_{2g} orbitals allows for the possibility of weak ferromagnetism in this *multi-band (or multi-orbital)* system. Such ferromagnetism may occur precisely in this intermediate-high carrier density regime, and is strongly dependent upon the microscopic details such as orbital splittings and the magnitude of inter-orbital Hund's coupling.

For our discussion, we will require a few particulars of the conduction band states in STO, which are well-established. The low lying octahedral t_{2g} crystal field levels of Ti comprise yz, xz, and xy orbitals. Owing to its directionality, hopping amongst xy orbitals occurs most strongly with neighbors in the x-y plane, and much more weakly in the z direction (a relative strength of approximately 1/8 is assumed for the ratio of z axis to x-y hopping in Ref. 12); the same considerations apply to yz and xz orbitals with appropriate rotations. Thus in a bulk system with cubic symmetry, there are three bands each of which disperses predominantly in 2 of the 3 cartesian directions. When confinement is introduced in the z direction for a (001) inteface, the states become quantized and bound in this direction, leading to two dimensional subbands. The xz and yz bands, which disperse in the z direction, suffer the greatest loss in kinetic energy, and are lifted somewhat in energy. The lowest energy subband is therefore expected to be of xy character, while in higher subbands the xz and yz states appear. The lowest xy subband disperses fairly uniformly in the two dimensional plane, while the higher xz and yz subbands become approximately *one-dimensional*, owing to the quenching of one of two of their main dispersion directions.

The reduction of the kinetic energy of the xz and yz subbands suggests we consider them for possible ferromagnetic polarization. We therefore adopt a minimal model with three two-dimensional sub-bands for the (001) interface, with the Hamiltonian $\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_I$, where the kinetic energy is

$$\mathcal{H}_0 = \sum_{\mathbf{k},\alpha} \frac{k_x^2 + k_y^2}{2m_0} d_{0\alpha}^\dagger(\mathbf{k}) d_{0\alpha}(\mathbf{k}) + \sum_{\mathbf{k},\alpha,i=x,y} \left(\Delta + \frac{k_i^2}{2m_i}\right) d_{i\alpha}^\dagger(\mathbf{k}) d_{i\alpha}(\mathbf{k}). \quad (1)$$

Here $d_{0\alpha}, d_{x\alpha}, d_{y\alpha}$ describe the xy, xz, and yz bands (with spin polarization $\alpha = \uparrow, \downarrow$), respectively, m_i is an effective mass, and Δ is the subband crystal field splitting. We have assumed a tetragonal crystal field symmetry for the (001) interface in Eq. (1), so $m_x = m_y$. Since we always consider the band bottom, this is equivalent to taking a tight binding model with hopping amplitude $t_i = 1/(2m_i a^2)$, where a is the lattice spacing. Here we make an approximation $t_0 \approx t_x = t_y \equiv t$ so that all the effective masses are equal. We take on-site interactions, of the form

$$\mathcal{H}_I = U \sum_{\mathbf{r},i} n_{i\uparrow}(\mathbf{r}) n_{i\downarrow}(\mathbf{r}) + U' \sum_{\mathbf{r},i\neq j} n_i(\mathbf{r}) n_j(\mathbf{r}) - J_H \sum_{\mathbf{r},i\neq j} \mathbf{S}_i(\mathbf{r}) \cdot \mathbf{S}_j(\mathbf{r}), \quad (2)$$

where $n_{i\alpha}(\mathbf{r}) = d_{i\alpha}^\dagger(\mathbf{r}) d_{i\alpha}(\mathbf{r})$, $n_i(\mathbf{r}) = \sum_\alpha n_{i\alpha}(\mathbf{r})$, and $\mathbf{S}_i(\mathbf{r}) = \frac{1}{2}\sum_{\alpha\beta} d_{i\alpha}^\dagger(\mathbf{r}) \boldsymbol{\sigma}_{\alpha\beta} d_{i\beta}(\mathbf{r})$. As usual, we expect the intra-orbital interaction Hubbard U to be the largest interaction, with the inter-orbital interaction U' and the Hund's coupling J_H rather smaller, $U'/U, J_H/U \lesssim 0.3$.

To analyze this Hamiltonian, we treat the U' and J_H as small (which they are, relative to U), and consider possible ferromagnetic instabilities they induce. With $U' = J_H = 0$, the Hamiltonian is decoupled to three single-band problems, and hence does not support ferromagnetism. What we require for the analysis is the susceptibility of each orbital subsystem, including the effects of strong on-site U. For the xy subband, which is two-dimensional, an on-site interaction U has little effect, and in the low density (per lattice site) limit studied here the interactions can be exactly treated by a standard T-matrix ladder summation. The result is simply a Fermi liquid with small Landau parameters, which can be neglected at the level of the present consideration. Neglecting the Fermi liquid correction, the spin susceptibility of the xy band per site is then $\chi_{2d} = ma^2/(2\pi) = 1/(4\pi t)$. For the xz and yz subbands, however, being one-dimensional, the effect of U is critical. As is well-known, an arbitrary strength repulsive interaction in one dimension transforms a Fermi liquid into a Luttinger liquid. The Luttinger liquid is described at low energy by bosonization, which decomposes the Hamiltonian of

the one dimensional system into charge and spin modes, with independent propagation velocities v_c and v_s, respectively, which generalize the Fermi velocity v_F. Notably, when the fractional electron occupation x_1 of a one-dimensional chain is small, $x_1 \ll 1$, the effect of interactions is strongly enhanced (this is a result of the fact that, in one dimension, an arbitrary repulsive scatterer becomes perfectly reflecting). As a consequence, the spin velocity is strongly suppressed. In the large U/t limit this can be obtained from the results of Shiba[19] for the susceptibility χ_{1d}, and the Luttinger liquid relation $\chi_{1d} = a/2\pi v_s$, which gives

$$v_s \approx Jax_1^2, \tag{3}$$

where $J = \pi^3 t^2/(3U)$, modeled as a chain with hopping t. We expect that Eq. (3) holds for arbitrary U/t and $x_1 \ll 1$, but with J becoming of order U for $U \ll t$. Notably, in the same limit, the charge velocity is much larger, $v_c \sim tax_1$. The smallness of the spin velocity indicates the strong suppression of spin exchange in this limit, and as a consequence a much reduced energy cost to polarize the electrons in the xz and yz bands.

Consider now the energy change of the system on polarizing the electrons. If the spin (per site) in the xy band is M_0 and that in the xz and yz bands is M_1, this is (per site)

$$\Delta E = \frac{1}{2\chi_{2d}} M_0^2 + \frac{2}{2\chi_{1d}} M_1^2 - J_H(2M_0M_1 + M_1^2) \tag{4}$$

The ground state is paramagnetic if this quadratic form is positive semi-definite, as it is for a small J_H. Conversely, a ferromagnetic solution with $M_0, M_1 \neq 0$ is favored when it becomes unbounded below, which occurs when $J_H(J_H + 1/(2\chi_{2d})) > 1/(2\chi_{1d}\chi_{2d})$. Using the forms of the one-dimensional and two-dimensional susceptibilities, this condition gives

$$x_1^2 < \frac{J_H}{2\pi J}\left(\frac{J_H}{2\pi t} + 1\right) \approx \frac{J_H}{2\pi J}. \tag{5}$$

This instability means M_0, M_1 will increase until Eq. (4) becomes invalid and stabilize the system, and the natural guess is that this occurs when the less occupied excited subbands become fully polarized. Remarkably, this occurs for *arbitrarily weak* Hund's coupling J_H, provided the filling of the upper xz and yz subbands is sufficiently small (see Eq. (5)), and of course non-zero. This gives a mechanism for ferromagnetism at *intermediate carrier density*, when the total density is near the critical value needed to just populate the xz and yz subbands, with magnetism disappearing both for smaller and larger carrier density.

It may appear that Luttinger liquid physics is essential to this above picture, a scenario that might seem overly exotic and restrictive. However, this is not the case. What is really required is simply an enhancement of the magnetic susceptibility of the upper xz and yz subbands when their occupation is small. The same tendency should be present even in the presence of weak dispersion normal to the direction of orbital extent, e.g. in the y direction for the xz orbital. This effect may be included in the model by adding a small t' hopping term, and would be expected to cause a crossover from Luttinger liquid behavior to Fermi liquid behavior at low energy. By continuity, for small t'/t, this Fermi liquid must have an enhanced spin susceptibility captured by a large Fermi liquid correction F_0^a. However, the eventual two-dimensionality induced by non-zero t' must prevent the divergence of the spin susceptibility of the xz and yz bands at small x_1, and if this effect is too large, the ferromagnetic instability may be entirely removed. To estimate this, we note that the one-dimensional magnetic susceptibility divergence is cut off when the Fermi energy of the xz and yz bands (measured from their minimum) is comparable to the hopping t', i.e. $tx_1^2 \sim t'$. The same condition describes the change from an open Fermi surface to an elliptical one. This gives the condition $t' \lesssim J_H t/J$ for the ferromagnetic phase to occur (we neglect numerical prefactors here due to the imprecision of the matching argument).

Interfaces	Symmetry	Local orbitals
(001)	4-fold rotation	xz, yz; xy
(110)	2-fold rotation	$\frac{1}{\sqrt{2}}(xz+yz)$; xy; $\frac{1}{\sqrt{2}}(xz-yz)$
(111)	3-fold rotation	$\frac{1}{\sqrt{3}}(xy + e^{i\frac{2\pi}{3}}yz + e^{-i\frac{2\pi}{3}}xz)$, $\frac{1}{\sqrt{3}}(xy + e^{-i\frac{2\pi}{3}}yz + e^{i\frac{2\pi}{3}}xz)$; $\frac{1}{\sqrt{3}}(xy+yz+xz)$

TABLE I. The relevant local crystal symmetries and local orbital states for different interfaces. ';' delimits the sets of locally degenerate orbital states. Note that there could be a small hybridization between $\frac{1}{\sqrt{2}}(xz+yz)$ and xy orbitals for the (110) interface.

For the (001) interface, it is the tetragonal crystal field environment and the quasi-two-dimensional geometry of TiO_2 layers that lead to the particular orbital and band structure discussed above. Recently the (110) and (111) LAO-STO interfaces have also been prepared experimentally[20, 21], and both interfaces appear to support STO electron gases, though the (111) interface is polar and the (110) is not. As listed in Tab. I, these two interfaces have different local crystal field environments and hence different local orbital configuration from the (001) interface. Can these two interfaces also support ferromagnetism – under ideal disorder-free conditions – at certain electron fillings?

As usual, the e_g doublets are always higher in energy and do not play any role. For the (110) interface, the three t_{2g} orbitals are splitted into three non-

degenerate orbitals, $\frac{1}{\sqrt{2}}(xz+yz)$, $\frac{1}{\sqrt{2}}(xz-yz)$ and xy. In the first approximation, the local hybridization between $\frac{1}{\sqrt{2}}(xz+yz)$ and xy orbitals may be neglected. When these two orbitals form bands, they are also quasi-one-dimensional just like xz and yz orbitals for the (001) interface. Hopping among $\frac{1}{\sqrt{2}}(xz+yz)$ ($/xy$) orbitals occurs most strongly with neighbors along z ($/[1\bar{1}0]$) lattice directions. The $\frac{1}{\sqrt{2}}(xz-yz)$ subband is two dimensional and its band bottom is the lowest among the three subbands. Due to the reduced symmetry of the (110) interface, the two upper quasi-one-dimensional subbands are split. Based on our above discussion of ferromagnetic instability for the (001) interface, we also expect emergent ferromagnetism for the (110) interface when the filling of the quasi-one-dimensional subband is sufficiently small. Because the two upper quasi-one-dimensional subbands are not degenerate, there may even exist two ferromagnetic regime as the electron filling of the two upper subband is increased. One should note that the discussion here assumes no hybridization between $\frac{1}{\sqrt{2}}(xz+yz)$ and xy orbitals. In reality, there are always small hybridization between these two orbitals. If this hybridization is very small (smaller than $\mathcal{O}(\sqrt{J_H/J})$), the resulting two-dimensional Fermi liquid should still have a large spin susceptibility and ferromagnetism can still be present.

For the (111) interface, although locally the crystal field splits three t_{2g} orbitals into one a_{1g} state, $\frac{1}{\sqrt{3}}(xy+yz+xz)$, and two e'_{2g} states, $\frac{1}{\sqrt{3}}(xy+e^{i\frac{2\pi}{3}}yz+e^{-i\frac{2\pi}{3}}xz)$ and $\frac{1}{\sqrt{3}}(xy+e^{-i\frac{2\pi}{3}}yz+e^{i\frac{2\pi}{3}}xz)$, the electron hopping strongly hybridizes three orbitals and leads to two-dimensional Fermi liquids. Therefore, our mechanism for ferromagnetism cannot predict a ferromagnetic state.

In the above, we have discussed possible itinerant ferromagnetism in two dimensional STO systems. Other theoretical works have instead proposed mechanisms relying on localized electron moments. While we believe that *Mott* localization of electrons near the interface should not occur for ideal structures, sufficient disorder and interactions together might create some truly localized moments. If the localized electron mechanisms are correct, we predict significant dependence of the ferromagnetism on disorder, and indeed that it should weaken as sample quality is improved. The itinerant mechanism discussed here has its own distinct predictions. For example, varying the electron concentration away from the critical density by tuning the back gate voltage may easily suppress the ferromagnetism, and ferromagnetism should be absent at the (111) interface; neither prediction applies for the local moment mechanism[22].

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